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A feasibility study of using cosmic ray muons to monitor supercritical CO_2 migration in geological formations

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Abstract

In geological carbon dioxide (CO_2) storage, possible leakage of supercritical CO_2 from underground storage is one of the main threats to defeat the climate goals of carbon sequestration. A feasibility study of using cosmic ray muons to monitor supercritical CO_2 migration in geological formations is carried out in this study, which is focused on improving the accuracy of numerical simulation. In the simulation, during the process of supercritical CO_2 migrating underground, both change in the density and the material composition of the underground storage were taken into consideration. A model of promising underground storage sites was established. Propagation of cosmic ray muons in the underground storage model was investigated by Monte Carlo simulation. The results showed that this method could detect 5% change in the supercritical CO_2 volume fraction in the storage model at depths of about 1km. This was deduced by investigating the sensitivity of the number of the cosmic ray muons penetrating the storage model to the change inside the model, which in practical case, was caused by CO_2 migration inside the underground storage.

Keywords: cosmic ray muon radiography; movement of supercritical CO_2; geological storage; saline aquifer; material composition; feasibility.

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Nomenclature

E_i	initial energy of muons
E_0	the rest energy of a relativistic muon when the kinetic energy is zero
P	density × length
Z	nuclear charge number
A	atom number
w_j	the mass weight of j element in a compound or mixture
n_j	the number of j element in a compound or mixture
θ	zenith angle of incident cosmic muons
H_{atm}	the altitude of production for muons with a trajectory at large angles
R_{Earth}	the Earth radius
L	muon path
$E^{cr}_{0,\pi}$	critical energy for pions to produce muons
$E^{cr}_{0,K}$	critical energy for kaons to produce muons
brine	an aquifer model composed of NaCl and H_2O, density=1.1g/cm^3
cross section	the occurring probability for one kind of interaction
standard rock	a model of the underground rock with Z/A=11/22, density=2.70 g/cm^3
supercritical CO_2	a kind of CO_2 state, with density=0.75g/cm^3 in this paper

1. Introduction

To alleviate the negative effects caused by an excess of CO_2 in the atmosphere, carbon capture and storage (CCS) has proven to be the only effective way to reduce CO_2 emission into the atmosphere on an industrial scale to date [1]. In this approach, supercritical CO_2 is injected into geological formations with sufficient porosity, permeability and storage capacity. Suitable storage locations are further required to be deeper than 800 m, where the ambient pressure is 80 times that of the atmosphere, high enough to enable CO_2 in supercritical state with the ambient temperature considered. And besides, overlying impermeable rock – the so-called "cap rock", should be present to prevent supercritical CO_2 upwards migration through the underground strata [2].

Deep saline aquifers are the most widely spread geological formations available to store supercritical CO_2. Once injected, supercritical CO_2 will displace and replace some of the salty water, and accumulate rapidly and largely in the injection zones under the cap rocks. Before its immobilization in the underground storage, supercritical CO_2 migrates both upwards and laterally from the injection zones [3]. And the main reasons for monitoring the subsequent performance of the storage sites after the injection of supercritical CO_2 are as follows. Firstly, if the injection pressure caused by large supercritical CO_2 accumulation in certain period exceeds the fracture pressure of the cap rocks, the seal layers will thus be damaged, creating leakage pathways. Secondly, supercritical CO_2 plume may leak into potable water aquifers and cause pollution. Thirdly, supercritical CO_2 may migrate to the areas near geological faults or abandoned old wells, posing a threat of leakage. To sum it up, to ensure both the safety and the efficiency of CCS, all CO_2 storage sites will need to be monitored so as to make prompt leakage alert and help adjust the simulation models for the supercritical CO_2 dispersion in the storage [4,5,6]. Many different kinds of geophysical monitoring techniques are available for potential use, and seismic monitoring is the most commonly used in CCS demonstration projects at present. However, the traditional ways are episode and expensive, and the resolution is far from satisfying for CCS monitoring [7], so it's necessary to proceed to find novel ways to optimize the monitoring effects and costs.

High-energy cosmic ray muon radiography has been employed in recent years to image the inner structures of the targeted geological objects with scales reaching hundreds and even thousands of meters [8]. Radiography was preliminarily introduced to distinguish different materials and densities, and map the material distribution inside the targeted objects. Its feasibility depends on the difference degree of the ray particles' energy loss in each material involved. With the same basis, in CCS monitoring, cosmic ray muons would be utilized to provide information on the change in the monitored storage area rather than map the supercritical CO_2 distribution. Previous work has preliminarily shown the possibility in [9]. Its simplified simulation consider change of mean density in the underground storage, neglecting that in material composition, which might have an impact on the results. This paper carried on the feasibility study with improved precision by taking into consideration the change in material composition as well. The monitoring principle is elaborated in the second section. The third section presents the simulation process and the results, with improved precision by taking into consideration the change in material composition, which show that this method could detect 5% change in the supercritical CO_2 volume fraction in the storage model at depths of about 1km.

2. Cosmic ray muon radiography

2.1. Principle of muon radiography for CCS monitoring

In the process of radiography, one ray beam is shot into a targeted object from a stable particle source and the penetrating ray particles are received by a detector. Interactions between the ray particles and the object will occur along the path, causing energy loss of the particles and consequent ray attenuation. When the radiation period is fixed, the attenuation totally depends on the material and the density of the object along the ray path. Adjust the incident angle of the ray beam, and obtain the corresponding attenuation in this direction. Following this way, the information of different parts of the object can be obtained.

In theory, the cross section of each interaction between the ray particles and different elements can be calculated in Quantum Field Theory, and thus average energy loss rate can be obtained. When the ray particles are muons, the average energy loss rate of them to cross a matter consisting of one element is as follows [10]:

$$\langle -\frac{dE}{dQ} \rangle = a(Z,\ A,\ E) + b(Z,\ A,\ E) \cdot E \tag{1}$$

The average energy loss rate given by (1) is expressed in MeV/ (g·cm^2). A (Z, A, E) is the electronic stopping power (energy loss rate) including ionization and excitation, and b (Z, A, E) is the total contribution of the energy loss rate caused by radiative interactions, including bremsstrahlung, pair production, and photonuclear interactions.

A compound or mixture is considered to be made up of pure elements, and the muon energy loss parameters in it can be calculated according to the following formulas [11]:

$$w_j = n_j A_j \Big/ \sum_k n_k A_k \tag{2}$$

$$\langle \frac{dE}{dQ} \rangle = \sum_j w_j \frac{dE}{dQ} \Big|_j \tag{3}$$

From (2) and (3), the stopping power parameters of the standard rock, brine and supercritical CO_2 for muons with different energies can be obtained in theory. Furthermore, by integrating (1), the average range of the muons in an object can be acquired:

$$-\int_{E_i}^{E_0} \frac{dE}{a(E)+b(E)E} = \int_L d\varrho \tag{4}$$

For muons, the stopping power parameters a (Z, A, E) and b (Z, A, E) vary slowly with muon energy E [10], and by an approximation neglecting their dependence on muon energy in the process of muon energy loss, ranges of muons with several energies in different materials are obtained according to (4). As is shown in Fig. 1, with the muons energy increases, the average range of muons in different materials increases. It also demonstrates that the stopping power is the strongest in standard rock, and the weakest in supercritical CO_2. Thus, it can be inferred that as the volume fraction of supercritical CO_2 increases, the stopping power of the mixture of standard rock, brine and supercritical CO_2 in the underground storage will decrease.

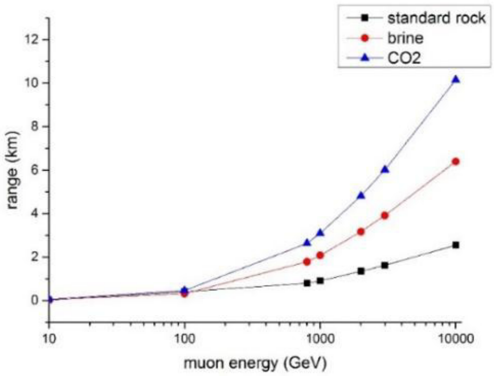

Fig. 1. The range of cosmic muons with different energies in the standard rock, brine and supercritical CO_2

2.2. Cosmic ray muons at the Earth's surface

The ground is continuously bombarded by cosmic ray muons with a nearly time-invariant energy spectrum, which is qualified to be a naturally occurring radiation source for CCS monitoring. There are several models for the cosmic ray muons energy spectrum at the sea level, and each of them has an energy range fitting well with the experimental data sets [12]. For this study, the modified Gaisser function was adopted. It is the most widely used model in projects employing high-energy cosmic muons to probe into the geological objects of large sizes. The Gaisser function for the energy distribution of cosmic ray muons flux from different zenith angles is as follows:

$$\Phi_G(E, \theta^*) = A_G E^{-\gamma}\left(\frac{1}{1+(E+\Delta E_0)\cos\theta^*/E_{0,\pi}^{cr}} + \frac{B_G}{1+(E+\Delta E_0)\cos\theta^*/E_{0,K}^{cr}} + r_c\right) \tag{5}$$

where

$$\cos\theta^* = \sqrt{1-\frac{1-\cos^2\theta}{(1+H_{atm}/R_{Earth})^2}} \tag{6}$$

In the modified Gaisser function, A_G is replaced by A_T. And amongst,

$$\Delta E_0 = 0.00206\left(\frac{1030}{\cos\theta^*} - 120\right) \tag{7}$$

The values for the parameters are given in Table 1, and corresponding to these values, the energy is expressed in GeV.

Table 1. Values of the parameters in the modified Gaisser's function

A_G	B_G	γ	$E_{0,\pi}^{cr}$	$E_{0,K}^{cr}$	r_c
0.175	0.037	2.72	103	810	10^{-4}

3. Geant4 Simulation

A model of promising underground storage sites was built, which was composed of an overlying standard rock layer of 1000 meters and a saline aquifer of 250 meters underneath the cap rock. The saline aquifer was the effective storage region for supercritical CO_2, consisting of brine and standard rock with a porosity of 35%. As supercritical CO_2 migrates after being injected, its volume fraction in the effective storage region changes. The sensitivity of comic ray muon radiography to this change needs to be investigated by Monte Carlo simulation, because each interaction involved during the muons propagation is stochastic with their respective probability in practice.

Geant4 is a Monte Carlo toolkit for simulating particles propagation through matter [13], which takes into account the dependence of the energy loss rate on the changing muons energy in the process beyond the reach of theoretical calculation. In the simulation, the detection area was 10000 m^2 and the detection period was set as one year. During one detection period, the supercritical CO_2 volume fraction was assumed to be stable in the targeted area to be detected. CO_2 volume fraction varied in separate measurement periods, so did the composition and the density of the saline aquifer of the underground storage. The outgoing number of the cosmic muons in each period was recorded by a muons detector beneath the underground storage adjacently. Due to the intrinsic fluctuation of the muons' behavior, the muons number detected in each detection period will have a range of variation around the theoretical average value in practical measurements, which approximately follows Gauss distribution [14].

The simulation results in Fig. 3 show that when the confidence level is chosen at 68.3%, the sensitivity for the change of the supercritical CO_2 volume fraction in the saline aquifer of the underground storage is 4%, and the sensitivity is about 5% when the confidence level is set at 90.0%. Here, supercritical CO_2 volume fraction change was taken as the relative change of the volume occupancy value between two

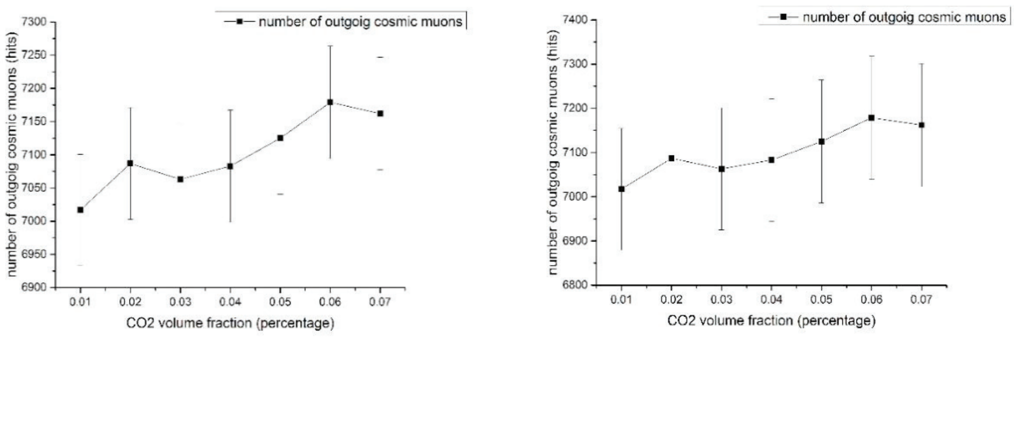

Fig. 2. The outgoing number of the cosmic muons under different volume fractions of supercritical CO_2 in the saline aquifer of the underground storage. Left: confidence level at 68.3%. Right: confidence level at 90.0%.

separate simulated conditions. The results preliminarily indicated a higher sensitivity of this method than the previous study.

4. Conclusion and outlook

The simulation took into account the influence of both the material composition and the density change caused by supercritical CO_2 volume fraction change in the underground storage, and thus, the feasibility study has been further certified with a higher accuracy, showing a higher resolution of the change in the underground storage. The sensitivity for the change of supercritical CO_2 volume fraction in the underground storage is about 5%, with a detected area of 10000 m^2 and a detection period of one year, which can be adjusted as long as the multiplication of them remains unchanged.

The sensitivity of this method improves with larger detector area and longer detection period, but they are confined in practical realization. In view of the fact that the number of cosmic muons decreases as it goes deeper in the underground, cosmic ray muon radiography is more suitable to be used in monitoring shallower sites. Future work will also involve developing a more complex model of the geological storage, which is closer to the real conditions and easier for cosmic muons to penetrate actually.

References

[1] Haszeldine RS. Carbon capture and storage: how green can black be? *Science* 2009;**325**:1647-1652, Sep 25.

[2] Birkholzer J, Cihan A, and Bandilla K. A tiered area-of-review framework for geologic carbon sequestration. *Greenhouse Gases: Science and Technology* 2014;**4**:20-35.

[3] Michael K, Allinson G, Golab A, Sharma S, and Shulakova V. CO_2 storage in saline aquifers II–Experience from existing storage operations. *Energy Procedia* 2009;**1**:1973-1980.

[4] Nordbotten JM, Celia M A, and Bachu S. Injection and Storage of CO_2 in Deep Saline Aquifers: Analytical Solution for CO_2 Plume Evolution During Injection. *Transport in Porous Media* 2005;**58**:339-360.

[5] Birkholzer JT and Zhou Q. Basin-scale hydrogeologic impacts of CO_2 storage: Capacity and regulatory implications. *International Journal of Greenhouse Gas Control* 2009;**3**:745-756.

[6] Lemieux JM. Review: The potential impact of underground geological storage of carbon dioxide in deep saline aquifers on shallow groundwater resources. *Hydrogeology Journal* 2011;**19**:757-778.

[7] Wiese B, Zimmer M, Nowak M, Pellizzari L, and Pilz P. Well-based hydraulic and geochemical monitoring of the above zone of the CO_2 reservoir at Ketzin, Germany. *Environmental Earth Sciences* 2013;**70**:3709-3726.

[8] Tanaka H, Nakano T, Takahashi S, Yoshida J, Takeo M, Oikawa J, *et al.* High resolution imaging in the inhomogeneous crust with cosmic-ray muon radiography: The density structure below the volcanic crater floor of Mt. Asama, Japan. *Earth and Planetary Science Letters* 2007;**263**:104-113.

[9] Kudryavtsev VA, Spooner NJC, Gluyas J, Fung C, and Coleman M. Monitoring subsurface CO_2 emplacement and security of storage using muon tomography. *International Journal of Greenhouse Gas Control* 2012;**11**:21-24.

[10] Gaisser T K and Stanev T. High-energy cosmic rays. *Nuclear Physics A* 2006;**777**:98-110.

[11] Groom D E, Mokhov N V, and Striganov S I. MUON STOPPING POWER AND RANGE TABLES 10 MeV–100 TeV. *Atomic Data and Nuclear Data Tables* 2001;**78**:183-356.

[12] Lesparre N, Gibert D, Marteau J, Déclais Y, Carbone D, and Galichet E. Geophysical muon imaging: feasibility and limits. *Geophysical Journal International* 2010;**183**:1348-1361.

[13] Agostinelli S, Allison J, Amako K, Apostolakis J, Araujo H, Arce P, *et al.* Geant4—a simulation toolkit. *Nuclear Instruments and Methods in Physics Research Section A* 2003;**506(3)**:250-303.

[14] Arslan H and Bektasoglu M. Geant4 Simulation Study of Deep Underground Muons: Vertical Intensity and Angular Distribution. *Advances in High Energy Physics* 2013;**2013**:1-4.